**Dear Reviewer A**

Thanks for your kindly attention. We have tried to answer your comments, meticulously. The sentences in the paper which address your precious comments are highlighted by using yellow pattern.

We hope that our answers can be enough clear, complete and acceptable. We are looking forward to receiving positive answers from you. Thanks for your kindly attention.

Truly Yours,

Mehdi Rashidzadeh

**Reviewer A:**   
**Is the manuscript clearly and concisely written?**   
**Ans.** Based on the reviewer comments, the manuscript was revised.

**Are the English grammar and syntax satisfactory?**  
**Ans.** The English grammar and syntax of manuscript was totally improved.

**ADDITIONAL COMMENTS**

**The authors answered that “the amount of acid sites that created on different calcined samples is not changed during NH3-TPD analysis”, but I commented that the amount of acid sites could be changed during thermal treatment before NH3-TPD analysis, having in mind that pretreatment was performed at temperature (500 ºC) higher than temperature of sample calcination (350 and 450 ºC). Although the sample was prepared by calcination at, for example, 350 ºC (and the characterization was performed for that sample), subsequent calcination at 500 ºC (before NH3-TPD analysis) will certainly change the structure of the sample. Although it “routine procedure in AutoChem 2900 (Micromeritics) automatic analyzer”, as author stated, the temperature of pretreatment should not be so high as to change the structure. I suggest repetition of the NH3-TPD analysis for samples calcinated at 350 and 450 ºC, but with lower temperatures of pretreatment (for example 300 ºC as for textural properties determination).**

**Ans.** Thanks for your attention. Due to the following reasons, it seems that pretreatment at a temperature higher than calcination temperature does not significantly affect the NH3-TPD results:

* Since pretreatment of samples is done under inert gas conditions (under helium flow), we expect that the amount of acid sites that created on different calcined samples is not changed during NH3-TPD analysis. This hypothesis is supported by the data in TABLE 2 of the manuscript. For example, pure SBA-350 and pure SBA-450 samples that experienced the same pretreatment temperature have different acidities for both weak and strong sites.
* According to the Referee's suggestion, acidity of SBA-350-30Al sample was measured whilst it was pretreated at 300 ºC instead of 500 ºC. The following table confirms that the results of NH3-TPD measurements of these samples have slight differences.

|  |  |  |  |
| --- | --- | --- | --- |
| Sample | Acid sites, µmolNH3g-1 | | |
| Weak | Strong | Total |
| SBA-350-30Al (Pretreated at 500 ºC) | 650 | 819 | 1469 |
| SBA-350-30Al (Pretreated at 300 ºC) | 662 | 840 | 1502 |

* Besides, pretreatment at a temperature higher than calcination temperature has also been reported in some other articles, such as:
* X. Zheng, B. Dong, Ch. Yuan, K. Zhang, X. Wang, *J Porous Mater* **20** (2013) 539 (DOI 10.1007/s10934-012-9626-6) "Direct synthesis, characterization and catalytic performance of Al-SBA-15 mesoporous catalysts with varying Si/Al molar ratios": Calcination at 550 ºC and NH3-TPD pretreatment at 600 ºC.
* [Zh. G. Wang](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-1), [J. N. Pei](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-2), [Sh. L. Chen](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-3), [Zh. Zhou](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-4), [G. M. Yuan](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-5), [Zh. Q. Wang](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-6), [G. Q. Ren](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-7), [H. J. Jiang](https://link.springer.com/article/10.1007/s12182-017-0150-9#auth-8), [*Petroleum Science*](https://link.springer.com/journal/12182) **14**,  (2017) 424 ([DOI 10.1007/s12182-017-0150-9](https://doi.org/10.1007/s12182-017-0150-9)) "The surface properties of aluminated meso–macroporous silica and its catalytic performance as hydrodesulfurization catalyst support": Calcination at 500 ºC and NH3-TPD pretreatment at 550 ºC.

**The authors answered that “Due to higher number of acid sites resulted due to higher amount of aluminum loading (NH3-TPD results), the SAXS analysis was performed only for SBA-450-(y) samples for investigating the effect of Al-loading on the structure of samples”. But, the reader can not know that at the beginning of the Results and discussion. It should be say, for example: “In Fig. 1 the SAXS patterns of SBA-450-y samples are presented, as examples, to analyze the effect of Al-loading on the structure of samples.” In addition, it would be useful to perform SAXS analysis for, for example, samples SBA-350-30Al and SBA-550-30Al in order to discuss the influence of calcination temperature on the structure.**

**Ans.** Thanks for your attention. Due to the higher number of acid sites resulted from the higher amount of aluminum loading, Based on NH3-TPD results, the SAXS analysis was performed only for SBA-450-(y) samples for investigating the effect of Al-loading on the structure of samples. According to your comment, the first sentence in SAXS section (Page 4) was revised and rewritten as follow:

“In Fig. 1, the SAXS patterns of SBA-450-y samples are presented, as examples, to analyze the effect of Al-loading on the structure of samples.”

**• The SAXS analysis showed that samples with Al have higher crystallinity than sample without Al, but the sample with higher content of Al had lower crystallinity than sample with lower content of Al. The same situation is for the positions of reflections. The authors avoided to discuss and explain such results. Please, give an explanation! If you use different Al quantity, you need to explain the influence of Al quantity on the samples structure.**

**Ans.** This comment will be answered with the next one.

**The authors used the results of Mouli et al. and Luan et al. works (references 13 and 10, respectively) to prove tetrahedral coordination of aluminum. The discussion should be more comprehensive, not just “The shift to lower 2θ value is well in line with presuming tetrahedral coordination of aluminum in the silica SBA-15 framework”. For example: “The shifting of the peaks to lower 2θ values can be indication of tetrahedral coordination of aluminum in the silica SBA-15 framework, as it was concluded by Mouli et al. and Luan et al. These authors proved tetrahedral coordination of aluminum by 27Al MAS NMR and concluded that shifting of 2Ѳ in SAXS diffraction patterns to lower values may be linked with longer Al–O bond than that of Si–O bond.” In addition, shifting of the peaks to higher 2Ѳ values with the increase of Al content should be explained.**

**Ans.** Your comment is acceptable. For better explanation, the corresponding sentence was modified in the manuscript (Pages 4&5) as below:

“In diffraction patterns of SBA-450-(y) samples, reflections of SBA-450-10Al and SBA-450-30Al have higher intensity compared to the pure one. Therefore, the degree of crystallinity is enhanced by grafting aluminum. Although SBA-450-10Al sample with high aluminum oxide content has lower crystallinity than SBA-450-30Al one. This phenomenon is probably due to the formation of aluminum islands in SBA-450-10Al sample, and consequently causes a decrease in the order of structure. As a significant observation, the SAXS analysis confirms that corresponding values for all three reflections (100, 110 and 200) shift to the lower 2*θ* values for SBA-450-10Al and SBA-450-30Al samples. Shifting these peaks to the lower 2*θ* values can indicate tetrahedral coordination of aluminum in the silica SBA-15 framework which is also concluded by Mouli et al. and Luan et al. by using 27Al MAS NMR.10,13 According to those studies, shifting of 2*θ* in SAXS diffraction patterns to lower values may be linked with longer Al–O bond than that of Si–O. Moreover, as seen from Fig. 1, positions of peaks in SBA-450-10Al and SBA-450-30-Al samples are not significantly different. This observation indicates that these samples probably have the same size of unit cell.”

**• Still not clear what it means “uniformity of pore size distribution”. Whether the “uniform pore size distribution” is the same as “narrow size distribution”? If it is, it can not be say, according to Fig. 3d that “the pore size distribution curves of SBA-350-10Al and SBA-350-30Al samples have less uniformity than that of the pure one”.**

**Ans.** Thanks for your attention. Uniformity refers to shape of pore size distribution (PSD) curves. As shown in Figs. 3(e) and 3(f), after grafting the aluminum, the PSD curves for samples that were calcined at 450 and 550 °C were approximately same as pure ones. However, for samples that were calcined at 350 °C, it was observed that the uniformity of the curve was less than pure SBA-350 sample, and the corresponding curves were not bell-type. Then, in our opinion, the uniformity is not as same as narrow pore size distribution. Therefore, for preventing any misunderstanding, the related sentence in the manuscript was omitted.

**• There is no AlO2 compound! There is AlO2– ion, which can form Al-oxide. Therefore, it should be stated: “probably due to filling their pores by Al-oxide species”, as it is stated in the introduction (One of disadvantages of post-synthesis method is the formation of oxides inside the mesopores that can partially or fully block them).**

**Ans.** Your comment is acceptable. The corresponding sentence was modified in the manuscript (Page 6) as below:

“Besides, N2 adsorption–desorption isotherm of SBA-x-(y) samples exhibits the reduction in the capillary condensation step of Al-containing samples probably due to filling their pores by Al-oxide species.”

**• The authors concluded at the end of the Results and discussion that “an increase in the amount of aluminum higher than Al/Si molar ratio of 0.033 establishes aluminum oxides in octahedral coordination, and this extra aluminum species does not affect acidity. Moreover, aluminum islands are created by increasing aluminum; therefore, total acidity of samples is reduced due to overlapping acidic sites.” Not aluminum islands, but aluminum oxide islands! This assumption can be related to the results of SAXS analysis and determination of textural characteristics.**

**Ans.** Thanks for your comment. The corresponding sentence was modified in the manuscript (Page 11) as below:

“Moreover, aluminum oxide islands are created by increasing aluminum; therefore, total acidity of samples is reduced due to overlapping acidic sites.”

These results are also supported by SAXS analysis and textural characteristics.